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Author(s): Dorhout, Jacquelyn Marie
Wilkerson, Marianne Perry

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A Review of Nuclear Forensics: 2016 – 2020

Jacquelyn M. Dorhout,^a Marianne P. Wilkerson^b

^aActinide Material Processing and Power Division, Los Alamos National Laboratory, Los Alamos NM 87545

^bChemistry Division, Los Alamos National Laboratory, Los Alamos NM 87545

Introduction

The study of nuclear forensics harkens back to the Manhattan Project-era, when scientists first started to analyze the debris from the 1945 Trinity test. Political turmoil stemming from the Cold War and the rehabilitation of Germany following WWII has led to new challenges in international security involving nuclear proliferation [1]. Nuclear materials have, on occasion, been lost, misplaced, or stolen from former Soviet countries, and illicit materials have been interdicted all over the world [2]. The National Technical Nuclear Forensics Center (NTNFC) was established in 2006, and has been at the forefront of drive to advance nuclear forensic capabilities in the United States [3]. The ultimate goal of nuclear forensics is to examine nuclear and other radioactive materials using analytical techniques to determine origin and history of the material, particularly in the context of law enforcement investigations [4,5].

Nuclear forensics can be divided into two parts: pre-detonation and post-detonation. Pre-detonation forensics, as the name implies, is the investigation of a nuclear material or weapon that has not been detonated or involved in an explosion, whereas post-detonation forensics is the study of activation or fission products in debris or the environment following the use of a nuclear or radiological dispersal device (RDD). Both parts require a number of analytical chemical and radiochemical techniques to determine identification of the material.

Many advancements in analytical techniques, including rapidity, sample size, and forensic signatures have been made in recent years. The analytical methods that can be used in a nuclear forensic investigation, such as mass spectrometry and gamma spectroscopy, have been described in detail in previous reviews, including Straub et.al. [6], and will not be explained here. This review will discuss recent publications (from 2016 to present) describing advancements of techniques such as

radiochronometry, morphology, development of novel reference materials, and inter-laboratory collaborations for both pre- and post-detonation nuclear forensics.

Pre-Detonation Forensics

1. Radiochronometry

Radiochronometry is the use of daughter (or granddaughter)/parent isotope ratios to determine the model age of a sample. The model age calculated for a sample assumes: 1) the parent is completely purified of daughter at the time of production and 2) the system has remained closed with no loss or gain of parent or daughter since production. Model ages can aid in the determination of sample provenance, the date of last separation of daughter products from the parent material (assuming a clean separation), or can support the verification of the source of starting material in nuclear production. Quantification of the daughter/parent isotope ratios used for radiochronometry can be done using several methods: thermal ionization mass spectrometry (TIMS), secondary ion mass spectrometry (SIMS), gamma ray spectrometry, alpha spectrometry, or inductively coupled plasma mass spectrometry (ICP-MS). TIMS and SIMS are most often used for particle age dating [7,8], whereas gamma spectrometry, alpha spectrometry, and ICP-MS are more often used for bulk measurements [6].

The most common radiochronometers applied to nuclear forensics are the decay of ^{234}U to ^{230}Th [9-11], and the progeny/parent pairings of $^{214}\text{Bi}/^{234}\text{U}$, $^{229}\text{Th}/^{233}\text{U}$, $^{228}\text{Th}/^{232}\text{Th}$, $^{234}\text{U}/^{238}\text{Pu}$, $^{235}\text{U}/^{239}\text{Pu}$, $^{236}\text{U}/^{240}\text{Pu}$, $^{231}\text{Pa}/^{235}\text{U}$, and $^{241}\text{Am}/^{241}\text{Pu}$. Previous reviews by Mayer [12], Keegan [13], Aggarwal [14], and Kristo [15] include extensive discussions on historical applications of uranium and plutonium radiochronometric signatures related to nuclear forensics published until 2016. Since then, a number of radiochronometry studies have been published on uranium [7-11,16-26], plutonium [21,27-36], protactinium age dating of uranium [37-47], other actinides and lanthanides [48-54], cesium [55,56], and strontium [57-60]. These updates to

radiochronometry, particularly the measurement of protactinium daughter products in uranium, are important advancements in the ability to determine the model age of an unknown material. The addition of new chronometers also advances the ability to interpret age signatures and investigate the behavior of nuclides used for radiochronometry. Discussions on the purification, characterization, and standardization of new tracer materials can be found in the Source Preparations section.

a. Uranium

The most common chronometer used in uranium age dating is $^{230}\text{Th}/^{234}\text{U}$ [61], although $^{229}\text{Th}/^{233}\text{U}$ and $^{214}\text{Bi}/^{234}\text{U}$ are used as well [61,62]. Recent radiochronometric studies often determine the daughter/parent ratios by single-collector (SC) or multi-collector (MC) ICP-MS [16,18,19,22-24,26]. Efforts have been taken to explore and compare the precision of SC vs. MC for age dating. Single collector instruments are less expensive and more readily available than MC instruments. Treinen et.al. published a study on uranium chronometry via single- and multi-collector mass spectrometry using certified reference materials (CRMs) CRM 125-A, U630, and IRMM-1000 and the $^{230}\text{Th}/^{234}\text{U}$ and $^{231}\text{Pa}/^{235}\text{U}$ chronometers [23]. The calculated ages of the CRMs were generally within a few months of the certified model dates.

Samples for ICP-MS, alpha spectrometry, and TIMS must be dissolved and purified prior to analysis, and isotope dilution methods are often utilized in order to quantify the concentrations of low level decay products. In some cases, non-destructive analysis (NDA) methods are the only means to determine the isotopic composition of a sample. Apostol et.al. used planar and coaxial gamma spectrometry to measure the uranium isotope ratio of $^{234}\text{U}/^{235}\text{U}$ in samples of metallic samples of non-regular geometry [17]. The activity ratios of $^{214}\text{Bi}/^{234}\text{U}$ and $^{235}\text{U}/^{238}\text{U}$ were also measured. The results were compared to Monte Carlo Simulations and were in very good agreement, with uncertainties less than 10%.

A novel NDA method to determine the age of a uranium sample is to use the uranium-helium chronometer, as demonstrated by Gates et.al.[20]. Though the technique is often applied to Earth and planetary sciences, it had never previously been used to date fuel cycle materials. A uranium metal sample of known provenance was heated and the helium gas was collected and analyzed with a noble gas mass spectrometer. Gates et.al. calculated the age of the

material to be approximately one year older than the known casting date, possibly because the helium was incompletely degassed from the melt during casting [20].

b. Plutonium

Plutonium in particular has been of interest because it is a byproduct formed from the irradiation of nuclear fuel, and is a weaponizable material that has the potential to be found out of regulatory control. When the ^{238}U fuel captures a neutron, it can be transmuted to ^{239}U , a short-lived ($t_{1/2} = 23.5$ min) uranium isotope that undergoes β decay to ^{239}Np ($t_{1/2} = 2.356$ d), which subsequently decays by β decay to ^{239}Pu ($t_{1/2} = 2.41\text{E}4$ y) [12]. A number of different isotopes of Pu can form if the fuel is left in the neutron flux long enough. Subsequent neutron captures on ^{239}Pu can form ^{240}Pu , ^{241}Pu , and ^{242}Pu . Any ^{235}U in the fuel can capture a neutron rather than fission and eventually form ^{238}Pu . The isotopes present and their concentrations are dependent upon a number of parameters, including neutron energy spectrum, initial fuel enrichment, neutron flux, and irradiation time. Aggarwall[31] and Wallenius et.al. have published studies on the origin determination of reactor-produced plutonium [12,63].

Some common chronometers for Pu age dating are $^{235}\text{U}/^{239}\text{Pu}$, $^{236}\text{U}/^{240}\text{Pu}$ [28-30], and $^{241}\text{Am}/^{241}\text{Pu}$ [30-32], although $^{234}\text{U}/^{238}\text{Pu}$ [27] and $^{238}\text{U}/^{242}\text{Pu}$ [28] are also used. The parent isotopes, ^{239}Pu and ^{240}Pu are the most abundant isotopes of Pu, which leads to measureable amounts of ^{235}U and ^{236}U , respectively, in samples. The $^{234}\text{U}/^{238}\text{Pu}$ chronometer is used less often due to the small natural abundance of ^{238}Pu . The $^{238}\text{U}/^{242}\text{Pu}$ chronometer is difficult to use because of both the low natural abundance of ^{242}Pu and the high natural abundance of ^{238}U .

Much like with uranium chronometers, the concentration of isotopes in a Pu sample are measured using gamma or alpha spectrometry, SIMS, TIMS, or ICP-MS. A current challenge with Pu radiochronometry is a complete lack of Pu reference materials with certified model ages. One recent study addressed this gap by measuring the ages of four Pu references materials (CRMs 136, 137, 138, and 126-A) through an interlaboratory comparison [35]. A number of different chronometers, including those listed above, were used by three different laboratories to date the Pu CRMs. The overall goal of the work was to identify if multiple labs could make independent measurements and provide consensus ages on Pu CRMs and the resulting consensus values were generally within the expanded uncertainties of the production date as is known [35]. The authors in this intercomparison study concluded that the isotope

standards can be used for quality control in Pu nuclear forensics radiochronometry applications.

c. Protactinium

In the past five years, a number of projects have focused on improving use of the $^{231}\text{Pa}/^{235}\text{U}$ radiochronometer to increase the number of radiochronometer pairs available for U age dating [37-44,46,47,64]. Although the ^{235}U daughter product, ^{231}Pa , has been used in radiochronometry for decades [65,66], Wallenius et.al. wrote the first open-literature study of the $^{231}\text{Pa}/^{235}\text{U}$ chronometer using both alpha counting and mass spectrometry in 2002 [67]. Eppich et.al. presented novel methods for ^{231}Pa assay determination solely by MC-CIP-MS in 2013 [68]. The 3rd Collaborative Materials Exercise (CMX), an international round-robin exercise designed to develop international collaboration in forensics, was the first such exercise to use $^{231}\text{Pa}/^{235}\text{U}$ chronometry [69]. The results of the exercise spurred an interest in further Pa chronometry development due to the observation that Pa/U and Th/U ages are concordant in powders purified for CRM production, but are discordant in metals [70].

The 4th CMX that occurred in 2015 included ^{231}Pa age-dating of the samples. Results from this large international exercise were published in 2018 [64]. Of the eleven laboratories that provided data for this exercise, only one lab applied the $^{231}\text{Pa}/^{235}\text{U}$ chronometer to date the sample using mass spectrometry procedures developed by Eppich et.al.[68]. The results of the $^{231}\text{Pa}/^{235}\text{U}$ and $^{230}\text{Th}/^{234}\text{U}$ chronometers showed consistency, validating the assumption that there was a purification event that quantitatively removed the Th and Pa from the U. It is not likely that there would be a process that only partially purifies Th and Pa in a proportion that retained concordancy of the chronometers. These results helped to confirm the utility of a $^{231}\text{Pa}/^{235}\text{U}$ chronometer.

In 2018, Treinen et.al. at Lawrence Livermore National Laboratory (LLNL) published an improved Pa spike calibration method for nuclear forensics in which they modified the ^{233}Pa spike preparation and calibration methods, improved sample purification chemistry, and refined data reduction algorithms [46]. The most significant improvement was the use of an in-house ^{231}Pa standard to calibrate ^{233}Pa spikes, rather than the standard complex chemical separation procedures needed to produce ^{231}Pa secular equilibrium reference solutions from geological matrices [66,68]. The new method required 2 days rather than the 2 weeks necessary for the previous calibration method. Two CRMs, U100 and IRMM-

1000, were age dated using the $^{231}\text{Pa}/^{235}\text{U}$ chronometer and the results were compared to the standard $^{230}\text{Th}/^{234}\text{U}$ model. Because of the short half-life of ^{233}Pa (26.967 ± 0.004 days) [71], it must be produced on an as-needed basis. At LLNL, the ^{233}Pa is produced by milking ^{237}Np with 10M HCl and 0.05 M HF [46]. The ^{233}Pa sample is calibrated using purified legacy ^{231}Pa material at LLNL. Three samples of CRM U100 were analyzed for both $^{231}\text{Pa}/^{235}\text{U}$ and $^{230}\text{Th}/^{234}\text{U}$ chronometers, and the results are presented in Figure 1.

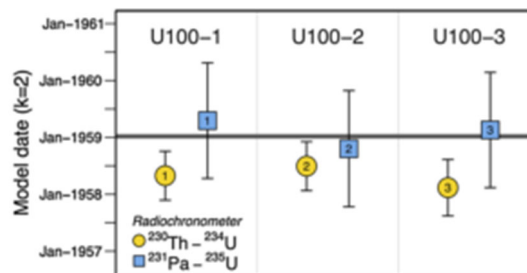


Figure 1. U100 results using both $^{230}\text{Th}/^{234}\text{U}$ and $^{231}\text{Pa}/^{235}\text{U}$ chronometers. All uncertainties reported are expanded uncertainties, $k=2$. Reprinted with permission from Treinen et.al.[46]

Both radiochronometry daughter-parent pairs agree with one another within uncertainty, although only the Pa/U chronometer preserves the material's production date of January 8, 1959; the Th/U chronometer indicates an older production date. The second CRM, IRMM-1000, was produced on July 9th, 2012 ± 13 days (see the Reference Materials section of Source Preparations) [24]. The $^{230}\text{Th}/^{234}\text{U}$ model has an average date of July 2nd, 2012 ± 28 days, which is in agreement with the known production history of IRMM-1000 [46]. However, the average $^{231}\text{Pa}/^{235}\text{U}$ date is ~ 70 days older than the average $^{230}\text{Th}/^{234}\text{U}$ date, with the two radiochronometers being discordant at the $k=2$ uncertainty level, an observation that has been reproduced from different IRMM-1000 stock solutions. Treinen et.al. conclude that Pa may have been incompletely separated from the IRMM-1000 CRM, but it is still a valuable quality control material. Additionally, they state that their study confirms the utility of calibrating ^{233}Pa using this new method, and demonstrates the need for a ^{231}Pa reference material for the radiochronometry community.

In 2017 and 2018, two international cooperation exercises took place wherein laboratories from multiple countries used the $^{231}\text{Pa}/^{235}\text{U}$ chronometer for age-dating uranium standards [39,44]. The results were published in 2020. Los Alamos National Laboratory (LANL) and LLNL in the United States

participated, as well as the China Institute of Atomic Energy (CIAE)[39] and the Japan Atomic Energy Agency (JAEA) [44]. LANL, LLNL, and CIAE each digested CRM U010 and U850 (uranium reference materials with known purification dates[72]) and measured the ^{231}Pa and ^{235}U assay by isotope dilution mass spectrometry (IDMS) using ^{233}Pa and ^{235}U isotope tracers. The purification dates calculated for both CRMs are generally concordant between the laboratories and agree with analytical uncertainty with the production history of both CRMs. Both CRMs represent good candidates for quality control during radiochronometry measurements of unknown materials for nuclear forensics.

In the second study, LANL, LLNL, and JAEA received samples of CRM U100, which was purified between December 3, 1958 and January 8, 1959 [72]. Model purification dates were estimated using the $^{231}\text{Pa}/^{235}\text{U}$ chronometer, similar to Denton et.al.[39]. The average purification date from all individual measurements is $\text{March } 26, 1959 \pm 237 \text{ days}$. This date agrees within analytical uncertainty of U100. Kayzar-Boggs et.al conclude by stating that the consensus model purification date of CRM U100 from this study may be used for quality control of future $^{231}\text{Pa}/^{235}\text{U}$ measurements of bulk low-enriched U materials [44].

The combined information from recent publications, including the 2016 review by Kristo [15], indicates that the use of $^{231}\text{Pa}/^{235}\text{U}$ is imperative for age-dating of uranium-containing materials for the purposes of nuclear forensics. As Kristo mentions, there is a global effort to develop multiple chronometers in order to expand the capability of material age-dating [15].

d. Summary

Radiochronometry has the ability to supply unique model age information that can support nuclear forensic assessments using a variety of chronometers ranging from ^{241}Am - ^{241}Pu and ^{230}Th - ^{234}U to ^{90}Y - ^{90}Sr and ^{137}Ba - ^{137}Cs . Though the chronometers are most accurate for nuclear materials that are highly purified, techniques such as alpha and gamma spectrometry can also be utilized to determine production dates in materials with more complex matrices. In the past few years, there have been more applications of radiochronometry, but there are still improvements needed in major areas, such as the availability of CRMs and their distribution internationally, the reproducibility of signatures using different techniques and materials, and more measurements and data to understand the behavior of parent and progeny nuclides throughout the U and Pu fuel and weapons cycles.

2. Morphology

The particle size, shape, density, and structure on the surface of materials can provide information on the processing history of the material, including precipitation conditions and calcination temperature; whether it was sintered or extruded; and sometimes the chemical composition of the material [73,74]. Previous reviews have covered morphology of nuclear forensic-related materials in general [12,14], particularly with ammonium diuranate (ADU) and ammonium uranyl carbonate (AUC) [12]. Generally, scanning electron microscopy (SEM) images are used to identify and compare the morphology of materials. Morphology has been used in previous experiments to compare the surface structure of ADU based on the ammonium source; typically ADU made with gaseous ammonia produces larger particles, while that made with aqueous ammonia tends to have less-well defined surface structures [75]. Morphology is useful for identifying material changes that occur from different process parameters, such as concentration, temperature, or stir rates [76]. Several studies have used morphology to compare precursor materials to products [77,78], and found that morphology is often retained through sintering. This information could be used to identify process conditions even in precursor materials. Similarly, morphology has been used to follow changes in material that have been stored under controlled conditions (i.e. temperature and humidity levels) over long periods of time [79]. These types of studies can help determine where material of interest had been stored (e.g. a climate-controlled laboratory, a dock or port) before it was interdicted. However, all of these morphology comparisons were done by describing the differences in the images using reasonable but non-quantitative language. Several techniques to identify and characterize the morphology of materials have been studied for nuclear forensics in the last few years, including two- and three-dimensional imaging with dual-beam focused ion-beam/scanning electron microscopy (FIB-SEM) [73,80], elemental mapping [81], and the use of the novel software Morphological Analysis for Materials (MAMA) [77-79,82-99].

a. Multi-dimensional Imaging

Chung et.al. have been focused on three-dimensional (3D) microstructural characterization of nuclear materials using a focused ion beam (FIB) technique combined with SEM [73,80]. Conventional two-dimensional (2D) imaging does not allow for obtaining depth profiles of the material, but 3D imaging can reveal inner microstructures. Both uranium and plutonium metal materials were analyzed using FIB-SEM, during which a sample was milled

using a Ga-ion beam source and then imaged in electron induced SEM mode (e-SEM) and ion beam induced SEM (i-SEM) mode [73]. Both modes show the preservation of material features using the Ga-ion beam milling compared to conventional milling techniques. Images can show inclusions from trace impurities, clearly defined grain boundaries, and pores within the sample. In addition, there is no etching or pitting present from milling. The sample is analyzed in slices in order to get the full volume picture [73].

A couple of years after the initial publication, Chung et.al published a follow-on study using FEB-SEM to characterize the microstructures within plutonium oxalate and oxide particles [80]. The internal morphology was analyzed to show the presence of internal pores and micro-cracks is much higher in plutonium oxide than in plutonium oxalate. The typical combined pore and micro-crack volumes are 1.9 and 5.2%, respectively. Additionally, the calcining of oxalate to form oxide does not change the outer morphology, but does contribute to the inner porosity due to the loss of gases during the calcining process. Further research is necessary to form a larger dataset of internal structures from various processing conditions, which could be used to quantify nuclear forensic signatures.

b. MAMA

Images from SEM analysis have been used for nuclear forensic analysis for many years [12,13,61,70,100,101], but it can often be a long and tedious process to identify various morphological indicators in databases containing hundreds of images [82,102]. Beginning in 2011, scientists at LANL worked to design a user-friendly software that could quantify particle sizes, and morphological characteristics such as shapes and surface particulate structures [82]. Ruggerio et.al. published an explanation of the software's interactive image segmentation and the algorithms it uses to learn from the operator input over time [84]. MAMA is designed to allow a user to analyze multiple particles in an image and calculate quantitative data such as number of particles, average pixel area, circularity of the particles, average diameter aspect ratio of the particles, and equivalent circular diameter (ECD) [87]. Figure 2 shows an example of an image that has been segmented using the MAMA software. The blue particles in the lower image were chosen by the user and analyzed for up to 22 attributes [90]. A full list of attributes and their descriptions can be found in Gaschen et.al. [87].

The first published use of MAMA was by Doyle et. al. in 2016 on a neptunium sample [86]. Neptunium oxide

morphology has been shown to change as a function of precipitation conditions and calcination temperature, and can often be correlated to a particular process. A sample of known NpO_2 was compared to an unknown sample using quantitative features calculated using MAMA including: number of particles, pixel area, ECD, ellipse perimeter, diameter aspect ratio, and circularity. The particles from the unknown sample were, on average, much larger and had a much larger ellipse perimeter than those from the known sample. The smaller area and ECD for the known sample correlates to documented atypical precipitation and calcination; higher calcination temperatures generally reduce the overall average particle size, meaning that the unknown sample was likely calcined at a higher temperature than the known sample [86]. These statistical values can be used to determine that the two samples did not come from the same process. When used in conjunction with other analytical methods, such as energy dispersive spectroscopy (EDS) and other wet chemistry methods [85], the MAMA analysis on the unknown material indicated that it likely originated from Savannah River National Laboratory in the United States [86].

The McDonald group out of the University of Utah has had multiple publications recently in which MAMA was used to analyze morphological features of nuclear materials [90-97,99,103,104]. They have analyzed and quantified the morphological features of U_3O_8 [90,96,97], uranyl peroxide decompositions [93], UO_3 [92,97], and uranyl oxide [94] and its decomposition [99]. The work from this group has allowed for the definition of the minimum number of particles that need to be analyzed in order to develop a statistically relevant assessment [90], and has also gone one step farther and introduced machine learning into the process in order to further simplify the data collection process for the user [92,95].

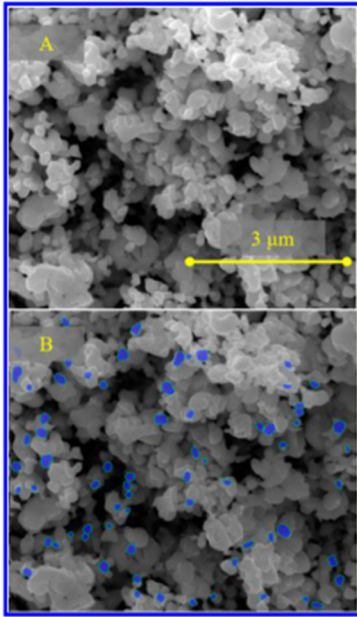


Figure 2. Example of an image before (A) and after (B) segmentation using MAMA. The scale bar is the same for both panels A and B. Reprinted with permission from Olsen et.al.[90].

c. Morphological Lexicon

After the development of a quantitative method to measure morphological features, it becomes necessary to accurately describe them qualitatively. Tamasi et.al. published a lexicon in 2016 which could be used to universally describe the morphology of a particle and its surface structure [89]. This lexicon has been cited numerous times in publications since [77,79,90,92,93,95,103-105]. It was built using terminology commonly utilized in geology, mineralogy, and crystallography [106], and was designed as a flowsheet of increasingly specific terminology. There are 11 steps in the lexicon, starting from whether the material consists of individual particles or clumped material, to whether the material is crystalline or rounded, to describing the surface of the material and if any processing effects (i.e. sintering, crushing, sieving) are apparent. In Figure 3, a high-level flow chart shows using the lexicon to describe the morphology of the material.

d. Statistical Analysis

Not only is MAMA being used to analyze materials in the laboratory, but those involved in statistical analysis are also interested in the data [83,98]. Anderson-Cook et.al. have been interested in designing experiments and identifying and analyzing signatures relevant to nuclear forensics for a number of years [83]. In a study designed to identify processing signatures, and

therefore processes, in PuO_2 , the sizes and shapes of particles were quantified using MAMA, and the lexicon was used to describe them. The manuscript outlined both statistical challenges as well as progress toward a comprehensive approach to certain nuclear forensics problems. In a second study, similar information was gathered for ADU materials [98]. In the case of both studies, the measured features, Y , and the inputs, X (i.e. experimental conditions) can be used together to determine the provenance of a material. For example, inferring about the X conditions that were used to create materials having specific characteristics, Y , is an inverse problem. Statistical analysis can be used to identify the best set(s) of X values for a set of observed Y responses. One can create a model based on known responses ($Y = f(X) + \text{error}$) and use it to “invert” and solve for X based on new based on new Y s. In a laboratory example, if it can be determined which quantitative values from MAMA are based on reaction conditions such as temperature or solution pH, a model can be constructed which should be able to give reaction conditions based on a new set of MAMA values. Reis’ paper in particular suggests that by using the functional inverse method, a higher predictive power can be achieved using fewer variables, allowing for streamlined and simplified data collection in the laboratory [98]. Both studies highlight that additional research is necessary into the impact of the model on inverse problems.

a. Summary

The use of morphology as a signature for nuclear forensics is not a new concept, but in the past few years, several new techniques have evolved that allow for more specific quantification of morphology. From imaging the internal structures of metals, oxalates, and oxides of the actinides, to utilizing the novel MAMA software to quantify the morphological differences between materials using statistical values, and defined universal lexicon descriptors, morphological analysis has been quickly advancing in recent years.

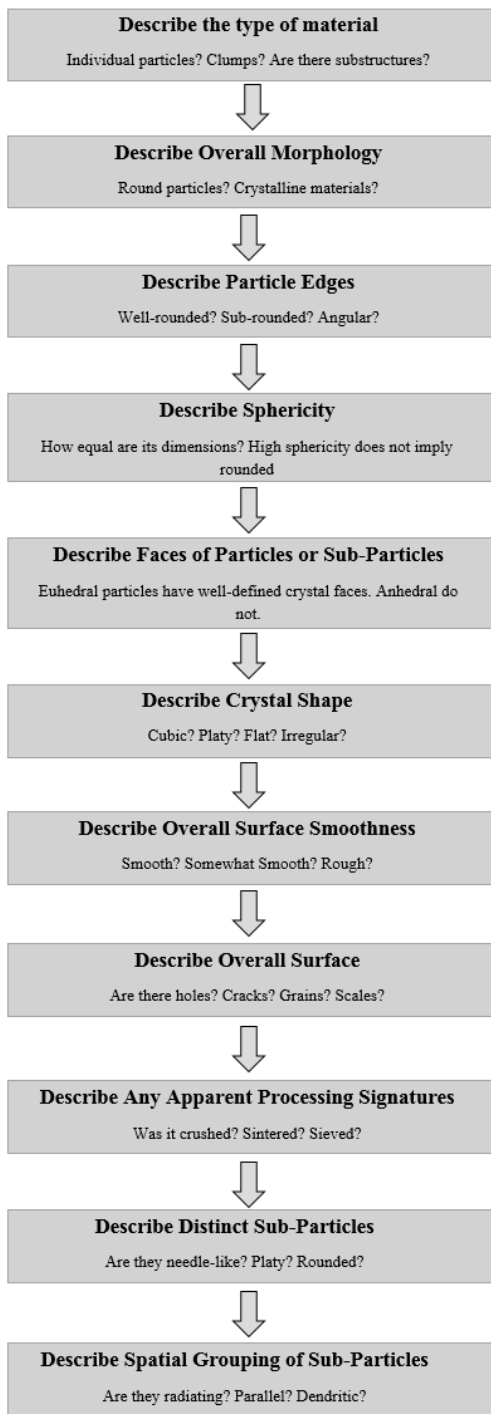


Figure 3. Using the lexicon as a flow chart to describe a material

3. Source Preparations

Using techniques like inductively coupled plasma mass spectrometry (ICP-MS), secondary ionization mass spectrometry (SIMS), or isotope dilution mass spectrometry (IDMS) cannot effectively be done

without proper standards and reference materials to which an unknown can be compared [107]. Reference materials are necessary to check the accuracy of results of an unknown, but are often only useful for measuring a major radionuclide with final purification dates that are assumed from the archives [25]. Certified reference materials (CRMs, also called standards) for radiochemistry have a known purification date and activity, an unbroken documentation of calibrations, measurements, and uncertainties, and are provided by an accredited institution (i.e. the Joint Research Centre of the European Commission, JRC-EC, the National Institute of Standards and Technology, NIST) [108]. A series of studies have been published in recent years focusing specifically on the production, isolation, characterization, and certification of various reference materials, tracers, surrogate materials, and standards.

a. Reference Materials i. Uranium

The radiochronometry model production date of a material is a predictive signature [25], meaning that it does not require comparison samples for assessment. However, quality assurance is of importance, and a reference material with a certified production date is necessary. Varga et.al prepared and characterized a novel uranium reference material that could be certified for production date based on the $^{230}\text{Th}/^{234}\text{U}$ chronometer [25]. Venchiarutti et.al. later certified the IRMM-1000a and IRMM-1000b materials in compliance with ISO Guide 34:2009 [11]. The reference material was produced as samples of dried uranyl nitrate with a known production date of July 9, 2012 at 11:08 am, at which point ^{230}Th was completely removed from the uranium nitrate material. Studies are on-going to certify the material for production date based on the $^{231}\text{Pa}/^{235}\text{U}$ chronometer [11]. Additional work on calibrations of reference materials was done by Chen et.al., who calibrated ^{229}Th for uranium age dating [18], and Mathew et.al. who expressed a need for Pu and U isotopic standards with lower uncertainties [21].

ii. Plutonium

IDMS is one method used to measure the isotopic abundances of Pu in a forensic sample. The most effective way to do so involves the use of a ^{244}Pu spike. Highly-enriched ^{244}Pu is not easily produced in reactors due to the necessity of long irradiation times and high neutron fluxes to achieve a significant proportion of ^{244}Pu . In the past 40 years, only two CRMs of ^{244}Pu have existed [109,110]. CRM 131 is obtained from the New Brunswick Laboratory (NBL) Program Office of United States Department of Energy (US DOE) and consists of 1 mg of plutonium

enriched to $0.97895\ n(^{244}\text{Pu})/n(\text{Pu})$ [109]. IRMM-042a is a nitric acid solution containing $1\ \mu\text{g}$ of $0.9793\ n(^{244}\text{Pu})/n(\text{Pu})$ plutonium, and can be purchased from the European Commission's Joint Research Center, Geel [110]. Essex et.al. point out that, while these CRMs are suitable for plutonium amount measurements, there are relatively high abundances of other Pu isotopes present, which limits the utility for some environmental samples because if a ^{244}Pu spike has a significant proportion of other plutonium isotopes, large uncertainties will result for the isotopic composition [33]. Using an enriched ^{244}Pu spike allows for a single mass spectrometric measurement that provides information on the amount of plutonium in a sample, and its isotopic composition. Typically, the isotopic composition analysis must be done separately. Additionally, Eppich, et.al. published a manuscript on plutonium assay and isotopic composition measurements in nuclear safeguard samples [111]. The majority of the discussion centers on the use of a ^{242}Pu spike to determine the concentrations of Pu in the samples by ICP-MS; however, the authors mention the use of ^{244}Pu as an alternative. A disadvantage of ^{242}Pu is that it cannot be reported independently, but is used as a spike to get measurements of the three most abundant isotopes (^{239}Pu , ^{240}Pu , and ^{241}Pu .) simultaneously in a single stage.

The IAEA recognized the need for a highly-enriched ^{244}Pu tracer, and in 1999 initiated a project to produce a plutonium isotope tracer with a ^{244}Pu enrichment greater than $0.999\ n(^{244}\text{Pu})/n(\text{Pu})$ [112]. Since the last major nuclear forensics reviews were published in 2016, a number of groups have been working to produce this tracer [33,34,36,111,113]. Penkin et.al. produced $0.88\ \text{mg}$ Pu with 99.98% ^{244}Pu purity using a two-stage electromagnetic separation of plutonium isotopes at the Russian Institute of Experimental Physics (VNIIEF) [36,114]. The initial sample was $0.5\ \text{g}$ PuO_2 roughly 17% enriched in ^{244}Pu . After the first stage, the purity was 98.8% . A second stage yielded a small sample of $99.983 \pm 0.001\ \text{at.}\%$ ^{244}Pu , which was verified by Lawrence Livermore National Laboratory (LLNL) in the United States in early 2020 [33]. Essex et.al. discuss in detail the stability, homogeneity, traceability (i.e. the result can be related to a reference through a documented unbroken chain of calibrations), verification, and measurement uncertainty of the ^{244}Pu reference material [33]. They conclude that the ^{244}Pu spike is a suitable IDMS reference material for use in measurements performed for nuclear forensics and nuclear safeguards.

iii. Protactinium

As discussed in the Radiochronometry section of this review, ^{231}Pa has become a radiochronometer of interest recently for age dating uranium samples [39,40,44,46,47,68,115]. However, a lack of reference materials has led to a number of publications on the production, purification, and use of ^{231}Pa and ^{233}Pa standards and tracers [37,41-43,45,46]. Jerome et.al. have been particularly focused on the production and purification of ^{231}Pa to be used as a standard [42,43]. As recently as 2020, they published results of an international collaboration to standardize ^{231}Pa as well as derive an updated half-life value [42]. The study included six laboratories from various countries, including the USA, France, Canada, and the EU, and resulted in good agreement on the activity with an overall uncertainty of 0.12% on the mean activity concentration [42].

Naperstkov et.al. published similar work on their method of production and standardization of a ^{233}Pa tracer in 2018 [45]. Although it will not be a commercially available ^{233}Pa reference material, it is another example of efforts to produce well-calibrated ^{231}Pa standards. Neutron activation of a thorium sample and subsequent purification using column chromatography produced ^{233}Pa , which was standardized for absolute activity using three independent methods. One sample was sent to the National Research Council (NRC) of Canada for standardization using $4\pi\beta\text{-}\gamma$ coincidence and anti-coincidence counting methods [116]. Two samples were counted using liquid scintillation counting methods from CIMEAT/NIST [117]. It was discovered that the ^{233}Pa sticks to the silanized glass ampoule, which makes it an unsuitable tracer for the calibration of an atom counting ^{231}Pa standard. Work is currently being done to stabilize the isotope in solution in order to make an appropriate tracer.

In 2019, Essex et.al. (LLNL) published a preparation and calibration of a ^{231}Pa reference material, which could be used to calibrate ^{233}Pa spikes [40]. Legacy ^{231}Pa material was purified and characterized by IDMS and MC-ICP-MS, and freshly separated ^{233}Pa spike solution was characterized by IDMS. The ^{231}Pa and ^{233}Pa samples were verified for massic radioactivity analysis as described previously [46]. Although the ^{231}Pa reference material was not considered a Certified Reference Material [118], the stability, homogeneity, and metrological traceability standards were met. Therefore, reliable ^{233}Pa spike calibrations can occur, which can be used to measure ^{231}Pa in $^{231}\text{U}/^{235}\text{U}$ model ages for nuclear forensics.

iv. Other Reference Materials

In addition to uranium, plutonium, and protactinium, tracer materials such as ^{229}Th [49,50], ^{243}Am [119], and ^7Be [120,121] have recently been produced and/or certified for use in nuclear forensics. The ^{229}Th is a reference material that has a certified thorium isotopic composition by mass (mol/g), rather than activity; characterization of which was done by several laboratories, including NIST, New Brunswick Laboratory (NBL), the Commissariat à l'énergie atomique et aux énergies alternatives (CEA) in France, and various national laboratories in the United States [50]. This reference is intended to be used for nuclear forensic analysis as an IDMS reference material. The studies determined that the reference is stable, homogenous, and has traceable, reproducible, and appropriate measurement uncertainties to be classified as a certified reference material.

Crozet et.al. filled another niche by conducting experiments to test a potential ^{243}Am reference material certified by mass (mol/g) rather than activity, a need that has been expressed by the International Atomic Energy Agency (IAEA) since 2009 to avoid error propagation from the half-life [119]. Both ^{241}Am and ^{243}Am can be used in radiochronometry to date Pu materials [119]. The CEA produced the initial material, which was 88% enriched in ^{243}Am , and distributed it to several laboratories, including laboratories in the United States, Germany, and France. The ^{243}Am content and isotope ratio of $^{241}\text{Am}/^{243}\text{Am}$ were among the data measured, and there was excellent agreement in the measurements between laboratories. Due to the success of the experiments, this ^{243}Am CRM is now available from JRC-Geel as IRMM-0243 [119].

Although beryllium is not an actinide, it is still of interest to the nuclear forensics community due to the use of ^6LiD in thermonuclear weapons [122], as the production of ^7Be is primarily done through the reaction $^6\text{Li}(d, n)$. Gharibyan et.al. produced and separated carrier-free ^7Be using proton bombardment of a $\text{LiOH}\cdot\text{Al}$ pellet with high yields and purity in 2016 [120]. The following year, Querfeld et.al. extracted low level activities of ^7Be from rainwater via evaporation and co-precipitation with $\text{Fe}(\text{OH})_3$, followed by several separation schemes and gamma spectrometry characterization [121]. Although their method needs to be optimized, and includes additional steps compared to Gharibyan [120], they showed that they could isolate ^7Be from rainwater with limited success, which could be improved upon to become a suitable low-cost tracer source [121].

b. Source Production

In addition to the production of reference materials and tracers, recent advancements have been made in the characterization and production of source materials, including the fabrication of surrogates [123,124], characterization of uranium particles [125,126], and analysis of Cf sources from commercial uses [127,128].

i. Surrogate Fabrication

Surrogate materials are often used in place of radioactive material debris. Foos et.al. synthesized glassy samples that were designed to be similar to tektite, which are complex glasses often formed during meteorite impacts, and generally contain large amounts of SiO_2 , Al_2O_3 , and FeO , among others [123]. Foos et.al. used sol-gel synthesis to form homogenous materials at lower than normal processing temperatures and then characterized their samples using thermogravimetric analysis, differential scanning calorimetry, XRD, and scanning electron microscopy (SEM) [123]. Though the tektite glasses were designed to be similar to natural occurring samples, heat treatment caused the formation of Fe and Ti-rich inclusions and crystallite formations, and thermal analysis showed the evaporation of volatile elements (i.e. chlorine). Using the sol-gel synthesis technique, the glasses can be doped with radioactive elements of interest during fabrication [123].

Another glass type, enstatite (MgSiO_3), was investigated by Reading et.al [124]. The glass beads are rapidly formed using a novel fusion method designed to produce homogenous, flux-free samples. Many elements can be measured using the glass bead method, but REEs are of particular interest to the nuclear forensics community. Originally designed to process geochemical materials, this method can be used to investigate silica-poor samples, including uranium ore concentrates or oxides, as enstatite is capable of dissolving complex materials and incorporating them into a glassy bead [124]. The samples were analyzed using LA-ICP-MS, which showed elemental homogeneity in the majority of the elements, as well as demonstrating volatilization of elements, such as Cu, Pb, Zn, and Sn, which was expected.

ii. Uranium Particle Characterization

The IAEA has been using trace samples to verify compliance of facilities around the world with international treaties, for which quality assurance is necessary [125]. Implementation of quality assurance requires characterized materials with particles as small as a few micro-meters, which are not readily available. Recently, Middendorp et.al. used a spray pyrolysis

technique to form aerosolized uranium particles of differing chemical compositions, which are then dried and characterized using scanning electron microscopy (SEM), energy dispersive X-ray spectroscopy (EDX), μ -X-ray diffraction (μ -XRD), and μ -Raman spectroscopy [125]. It was determined that the chemical composition of the particle affects the morphology, and an increase in production temperature often leads to an increase of the diameter of the particles, due to agglomeration. Additionally, the source material determines the isotopic composition of the final particle, and adjustment of the nonvolatile components can affect the size and elemental content of the particles [125].

A year later, Middendorp et.al. published a second study on uranium micro-particle suspensions using uranium oxide aerosols in order to create a characterized uranium oxide material for quality assurance [126]. The particles were characterized with the same techniques listed above, with the addition of quadrupole ICP-MS (Q-ICP-MS). The micro-particles were contacted with ethanol for more than one year, and the suspended particles were separated using a 0.45 μ m filter. Both the dissolved and suspended particles were analyzed by Q-ICP-MS to identify concentration; the filtrate solution was close to the 1 μ g limit of detection. Preparation of sample matrices based on real-life sampling materials (i.e. silicon wafers, cotton swipes) showed a reasonably large number of particles distributed homogeneously over the substrate, particularly when the uranium oxide particles were combined with cerium (IV) oxide (a plutonium surrogate) [126].

Both of these experiments show the possibility for the use of uranium micro-particle aerosols as quality assurance materials for IAEA trace samples. The technique has been proven to suitable for preparation of different types of reference materials prepared from particle suspensions.

iii. Analysis of Cf sources

Californium sources are often used in medicine, geology, nuclear safeguards, and industry, and they are produced almost exclusively by two facilities: Oak Ridge National Laboratory in the United States, and the Research Institute of Atomic Reactors in the Russian Federation [127]. Apostol et.al. recently published a study on the parameters of Cf neutron sources relevant to nuclear forensics in order to aid in investigations when Cf sources are found outside of regulatory control [127]. The study involved age-dating sources, isotopic composition analysis, identification of impurities, and non-destructive gamma spectrometric analysis. The isotopes ^{249}Cf and

^{251}Cf can be measured directly via gamma spectrometry, and the other two main isotopes, ^{250}Cf and ^{252}Cf , are estimated from the gamma lines of their fission products: the ratio of ^{137}Cs to ^{132}I [128-130]. Five sources with known activity dates were analyzed using a coaxial HPGe detector and Monte Carlo modeling, and the measured activities match well with their certifications [127]. Interestingly, the presence of ^{241}Am and ^{154}Eu was observed in the sources produced in the Russian Federation, but not in those produced in the United States. Neither isotope is a fission product of Cf, so the authors concluded that their presence is indicative of contamination rather than an incomplete separation. The isotope ^{154}Eu is used in control rods produced at the Research Institute of Atomic Reactors, and could account for the contamination [127].

Similarly, Gregor et.al. investigated non-destructive gamma analysis of Cf sources for the purposes of forensic investigations [128]. Four Cf sources were analyzed using a coaxial HPGe Falcon 5000 detector. The fission products of ^{252}Cf were identified, particularly ^{137}Cs . The ^{250}Cf has a weak gamma signal at 42.9 keV, and is more abundant in older sources due to its longer half-life (13.1 years vs. 2.65 for ^{252}Cf) [131]. Again, ^{154}Eu was detected in the gamma spectra of sources from the former Soviet Union or the UK, but not in those sources produced in the US [128].

The information obtained from these experiments could prove invaluable during nuclear forensic investigations, and lead to the discovery of material age and origin of Cf sources.

c. Summary

The preparation of reference materials and standards, and the characterization of Cf sources are instrumental in the accuracy of analysis in nuclear forensics. Within the past five years, several novel reference materials have been separated, isolated, and characterized for use in forensic analysis, including thorium, uranium, plutonium, protactinium, beryllium and americium. The creation and analysis of micro-particles of uranium and cerium for IAEA quality assurance has also been a recent advancement in forensic investigation. Finally, the characterization of known Cf sources, from age dating to impurity concentration, can be used to identify materials in investigations of Cf material out of regulatory control.

4. Exercises

The International Technical Working Group (ITWG) conducts a number of international exercises to keep the skills of the laboratories around the world in good practice. Two major types of exercises have been put on by the ITWG: the Collaborative Materials Exercise

(CMX), which often features samples of unknown composition and origin, and Galaxy Serpent, which was initiated to raise awareness of creating and using a national nuclear forensics library. The first three CMXs were held from 1999 to 2011, and are discussed in previous reviews [13,15].

a. CMX-4 through CMX-6

CMX-4 occurred in 2015, and the results of the exercise were published from 2017-2019 [64,132-139]. It involved 16 countries or international organizations. Exercise samples were shipped as part of an illicit trafficking scenario, and each laboratory was asked to conduct nuclear forensics analysis in support of a fictitious criminal investigation [133]. Over 30 analytical techniques were applied, including 10 techniques novel to CMX. Three low-enriched uranium oxide samples were distributed: one powder sample (ES-1) and two pressed and sintered oxide pellets (ES-2 and ES-3). Both sintered pellets originated from the same enrichment and fabrication facility with identical production specifications, but were created from separate batches roughly three years apart. Analytical techniques such as weight, dimensional analysis, pycnometry (density measurement), scanning electron microscopy (SEM), light optical microscopy, SEM backscatter diffraction, X-ray radiography, atomic force microscopy, Raman spectroscopy, powder X-ray diffraction (XRD), thermogravimetric analysis (TGA), radiochronometry, inductively coupled plasma – mass spectrometry (ICP-MS), ion chromatography, secondary-ion mass spectrometry (SIMS), and thermal ionization mass spectrometry (TIMS) were among those utilized. Age dating of the samples concluded that ES-2 was made at a different time than either ES-1 or ES-3, and that the ages of ES-1 and ES-3 are not significantly different. Isotopic compositions show that ES-1 and ES-3 were downblended with freshly enriched uranium that had not seen a reactor, but the batch used to generate ES-2 was significantly different. The majority of the participating laboratories were able to correctly identify the differences between ES-1, ES-2 and ES-3.

CMX-5 took place in 2017, and results from that exercise are now being published [140]. Laboratories from 19 countries and one multinational organization participated, and over 30 analytical techniques were used to investigate the samples. For this exercise, laboratories were tasked with determining where and when two materials left regulatory control, and locate security vulnerabilities within nuclear facilities. This was accomplished using group inclusion/exclusion analysis, in which the material in question is compared against standard material and nuclear forensic library

data. The goal was to improve international technical capabilities, cooperation, and communication. Analytical methods previously listed for CMX-4 were also used in CMX-5; Particle Induced X-ray Emission (PIXE) and porosity determinations were used for the first time. In the scenario, a radioactive object was discovered by authorities as it was being driven between two countries. Previously, a similar package had been seized after setting off a radiation monitor. Traces of uranium oxide were found, and the objects are suspected to be uranium fuel pellets. The two materials provided to the laboratories were high fired, low enriched uranium oxide pellets manufactured by the French Commissariat à l’Energie Atomique (CEA) with slightly different procedures for the purposes of this exercise in 2014. Results across the laboratories for age dating were consistent and correctly identified the age of the samples. However, state-of-practice isotopic techniques were not able to conclusively include or exclude samples due to the material isotopic similarity and identified that characteristics useful for inclusion/exclusion assessments are case specific and directly tied to the materials and the questions being asked by investigators. This strengthens the need to continue these collaborative material exchanges to develop case-by-case analysis methods.

CMX-6 was completed in 2019 and was based on a realistic scenario designed to exercise laboratory capabilities to test material out of regulatory control and provide technical solutions to national or international authorities [141]. Two radioactive samples were seized from a fictional metal recycling plant: a cerium metal block and a depleted uranium metal block, both of which were contaminated with plutonium fluoride. Both non-destructive and destructive techniques were used for analysis, similar to the previously described exercises. Results from the exercise should be published in the coming years.

b. Galaxy Serpent

In addition to strengthening laboratory techniques and practices, the nuclear forensics community has focused on building and utilizing a national library of signatures. Galaxy Serpent is a first-of-its-kind, virtual, web-based international tabletop exercise where teams of scientists from various countries use provided public domain data to compile their own library, and determine if hypothetically seized samples are or are not consistent with their library [142]. The addition of experts from a variety of fields, such as nuclear reactor engineers or fuel experts, helps increase the range of expertise in the nuclear forensics community. Three exercises have taken place since 2014 [142-144].

The first Galaxy Serpent exercise took place in 2014 and was comprised of two phases. In the first phase, participants were asked to organize a model library using provided spent fuel characteristics from three reactors. In the second, participants needed to determine if data from a hypothetical seizure is or is not consistent with a reactor in their model library [142]. Participating teams were able to successfully report identification of the likely reactor from which the hypothetical sample originated, as well as a set of “possibles”.

The second Galaxy Serpent exercise occurred in 2016 and focused on sealed radioactive sources as the material of interest [143]. This exercise consisted of three phases and involved 35 teams of scientists from 28 countries and three international organizations. In Phase 1, teams were given synthetic sealed source data developed by Argonne National Laboratory in the United States, which was used to compile a model library. In Phases 2 and 3, this library was used to compare whether simulated data from two fictitious scenarios were consistent with their library. Phase 2 was based upon synthetic data from a hypothetical intact sealed source found out of regulatory control. Teams were to make warranted assumptions and down-select to four potential matches in their model inventory. Phase 3 used simulated data from a hypothetical detonated radiological dispersal device. In this case, teams were to down-select to five potential matches in their inventory. Generally, teams offered reasonable assumptions and well-qualified down selection paths. Although teams differed in the number of potential matches reported, their choices were supported by defensible criteria.

Results from the third Galaxy Serpent exercise were published in 2019 [144]. In this version, the material of interest was a series of uranium ore concentrates (UOCs). A surrogate data set for the forensic signatures of UOCs was prepared based on real trace element data while preserving sensitive and proprietary information of existing datasets. As in previous exercises, in Phase 1, teams were given data on the four main classes of UOCs, which included 822 records, and were told to create a forensic library. The hypothetical scenario involved the recovery of barrels of UOC outside of regulatory control; one sample was designed to fit within the dataset, one sample was designed to be an outlier, and one sample was designed to be not represented within the dataset, providing additional challenges. In Phase 2a, teams were asked to determine if the materials were consistent with each other, and in 2b, whether the materials were consistent with any of the four classes of UOCs for which they had data. Teams successfully determined that two of the samples shared a common provenance but were not identical, and that the third sample was entirely

different. All of the teams were also able to correctly connect the two similar materials to one class in their dataset, but many struggled with the sample that was not represented in the given dataset. This shows that, while these libraries play a vital role in investigative efforts to identify unknown materials, the libraries are constantly growing and changing to accommodate new information. A fourth version of Galaxy Serpent is under development and was scheduled to take place in early 2020.

c. Summary

International exercises that involve multiple laboratories around the world are necessary for improving the capabilities in nuclear forensics. They allow for cooperation between laboratories and practice in the event of a real-world incident. Exercises like Galaxy Serpent show the need for a constantly updating library of forensic information to which real-world samples can be compared.

5. Simulations

As is true in most scientific fields, not all of the recent studies in nuclear forensics are based in a laboratory. In some cases, using a simulated data can give information that is not feasible to gather from an experiment, such as identifying unknown spent nuclear fuel [145]. There are a number of teams around the world who are interested in designing simulations of nuclear events [146] or buried sources [147], applying multiple statistical methods to collected data [83,98,148-152], developing new forensic models [153] or algorithms [154], optimizing databases [155-161], generating theoretical data sets [162-168], and utilizing machine learning techniques [169], all for the purpose of advancing nuclear forensics.

a. Computational Calculations

Some studies, such as Egnatuk et.al., use computational simulations to evaluate data that would be difficult to procure experimentally. They wanted to identify the gamma-ray spectra from 0.1 hours to 10 days after a fission event, and evaluate the activity of fission products in certain mass chains in order to differentiate the fission materials used in the event [162]. Their model assumed the presence of ^{239}Pu or ^{235}U in a spectrum of fission-energy neutrons, and the use of a widely commercially available HPGe (High Purity Germanium) detector to detect the gamma rays. An MCNP5 (Monte Carlo N-Particle transport code) model was used to generate gamma-ray spectra from each fissionable material, and assumptions were made on the number of fission events that occurred. In addition, the computational data can allow for the analysis of volatile and semi-volatile radionuclides, which may not be easily measured in experimental

samples. The results showed a distinguishable difference in the production of ^{89}Zr activity: none is produced when the fissioned material is ^{235}U , but a small, measurable amount is created from the fission of ^{239}Pu [162]. Additional differences in activity can be seen for ^{131}Te , ^{131}Sb , ^{132}Cs , ^{134}Cs , and ^{136}Cs , depending on the initial material. These computational results show that gamma-ray spectra, focusing on mass chains with volatile or semi-volatile radionuclides present, could prove useful for determining the initial fissile material in an event.

In a similar study, Redd et.al. used computational data to characterize nuclear material based on reactor operating characteristics and Pu quality, with the eventual goal of characterizing the by-products of a weapon detonation [166]. Monte Carlo simulations were used to calculate uncertainties in nuclear forensics signatures and production estimates (i.e. $^{242}\text{Pu}/^{240}\text{Pu}$ and $^{240}\text{Pu}/^{239}\text{Pu}$). A more thorough understanding of these uncertainties may allow for improvement of nuclear data and thus a stronger differentiation between production reactor designs, including core configurations [166].

Computational studies have also been used to determine neutron capture cross-sections on short-lived isotopes. Liddick et.al. used the β -Oslo method to extract the neutron capture cross-section of ^{50}Ti [165]. Their results demonstrate the validity of this recently-developed method, and provide a sufficiently small uncertainty in the data.

Comparisons of experimental results to computational data are also of interest. This concept may not be new, but noteworthy studies have been published in recent years by Goodell et.al., who compared irradiated foil measurements to HPGe simulations [163], Roach, et.al., who compared isotopic measurements of ultra-trace fission products to an ORIGEN model [167], and Usman et.al., who fingerprinted commercial nuclear reactors using ORIGEN simulations [168].

b. Database Development

Nuclear forensic investigations often depend on comparative signatures; those which rely on the comparison of characteristics to a set of known samples [155]. These databases can include a variety of information, such as spent nuclear fuel characteristics [154,155,157] or reactor type discrimination [156,158-161]. Su et.al. used a database to identify unknown spent nuclear fuel, including the reactor type, initial enrichment of the fuel, and the burn-up [155]. The database was initially constructed using simulated data, and was then optimized using real-world commercial information in

order to better meet the need of real scenarios. Information in the database was sourced from reactors all over the world, including the UK, USA, Canada, Germany, Japan, and Russia. Both the simulated and real-world data were used to identify three “unknown” spent nuclear fuel samples by comparing fuel enrichment and burn-up using linear regression analysis, which identifies which factors are related to concerns and estimates the relationship between dependent and independent variables; and linear discriminant analysis, which is a method used to determine the type of sample by grouping known data by type and calculating the linear discriminant function into which the unknown data can be substituted. Though there were limitations to using this optimized database, Su and coworkers conclude that the relative error decreased when using the optimized version compared to the version containing only simulated data, and they plan to continue improving the database for future scenarios [155].

While groups such as Su are interested in looking at spent fuel as a whole, others, such as Kitcher et.al. are focused specifically on the characterization of separated weapons-grade plutonium [156]. Their methodology also uses database comparisons, but the focus is on isotope concentrations that are produced in a reactor core. A library of plutonium and contaminant fission produce isotope ratios and burnup simulations produced from Monte Carlo calculations were used to determine a likely reactor type, fuel burnup, and time-since irradiation for an “unknown” sample. The study determined that accurate reactor core physics and burnup simulations are important because the uncertainties in determining reactor type are sensitive. Fuel burnup and time-since irradiation are comparatively invariable. The reactor simulations using the current (2019) database gave results with which the authors could identify limits on uncertainties on reactor-type discrimination. One test material could be identified at the 99% confidence level with 28% uncertainty, but the other could only be identified at the 95% confidence level with 14% uncertainty. Future simulations will look to minimize uncertainty, particularly within the Sm and Pu ratios in order to improve predicted burnup values [156].

c. Summary

The study of real-world nuclear materials isn’t always possible for a number of reasons, but simulations can be done to gather data and make reasonable estimates. The development of new models and the application of multivariate analysis methods in new ways can advance the study of nuclear forensics by optimizing databases and libraries and improving reactor simulations.

6. Case Studies

Casework in nuclear forensics is important because it highlights research and development in the field on real-world samples. Previous reviews have discussed a number of international seizures, historical material analysis, and international exercises from the early 1990s through 2015 [12-15]. The 2016 review by Kristo et.al. has a particularly in-depth analysis of the international seizures and laboratory exercises during this time period [15], and they will not be repeated here.

Recent case studies have included everything from investigating legacy samples with unknown provenance [170-173] and source identification of material [31,174-177], to analysis of nuclear fuel pellets [178,179] and measurement of fission and activation products in lichen materials [180]. Additionally, publications have appeared on the status of various nuclear facilities around the world, from Brisbane to Uzbekistan [181-187]. The following are just a couple of the numerous examples of interesting case studies on materials with unknown provenance, which demonstrates the ability of forensics techniques to be used to answer questions on a smaller scale.

a. PuO_2 with no Pedigree

In mid-2015, Norman et.al.[172] published a case study on a macroscopic sample of PuO_2 that had supposedly been purified by Cunningham and Werner on September 10, 1942 from a sample produced by Seaborg and co-workers (Figure 4) [188]. There are no written records of the material, but it was on display for a number of years at UC Berkeley. In 2008, it was offered to the Smithsonian museum due to its significance. The Smithsonian expressed concern over the authenticity of the sample, due to the lack of a paper trail, which led scientists at UC Berkeley to attempt to establish its authenticity. Non-destructive HPGe analysis was performed on the sample and showed gamma rays attributed to the decay of ^{239}Pu and those produced from the decay of ^{241}Am . The ratio of $^{241}\text{Am}/^{239}\text{Pu}$ was less than 2.3×10^{-7} , revealing that the sample was most likely produced from a low-neutron-flux environment rather than a nuclear reactor [172], which is consistent with Seaborg's description of the production of plutonium [189].

The mass of the sample was determined using efficiency calibrations of the detector and attenuation due to the plastic box containing the sample. The mass of ^{239}Pu in the sample was calculated to be $2.0 \pm 0.3 \mu\text{g}$, which is reasonably consistent with the original mass ($2.44 \mu\text{g}$ ^{239}Pu) of the sample produced by Cunningham and Werner [188]. Norman et.al.

conclude by stating that while the results are not 100% conclusive, non-destructive testing of the sample has shown that it is consistent with that prepared by Seaborg and his collaborators [172].

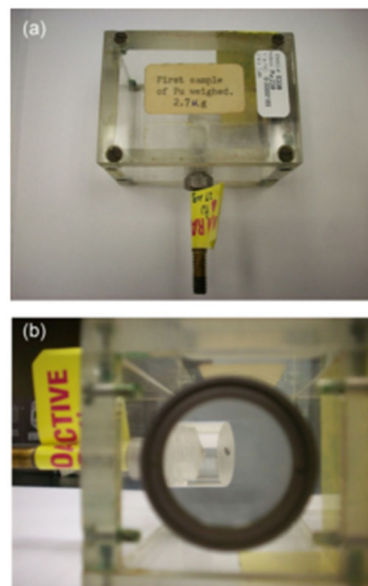


Figure 4. a) Outside of sample box with labels; b) side view showing sample attached to plastic rod. Reproduced with permission from Norman et.al.[172]

b. Unknown Inventory Sample

Researchers at Argonne National Laboratory in the United States published a similar study in 2017 [190]. A planchet containing Pu material was discovered during a de-inventory exercise whose origins could not be verified. Savina et.al analyzed the planchet using analytical techniques common to nuclear forensics in order to understand its composition and origin. Given the potential historical significance of the sample, only non-destructive techniques were used [190].

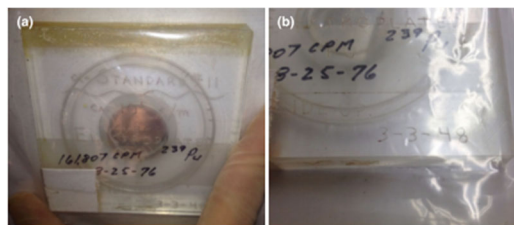


Figure 5. Photos of the ^{239}Pu planchet discovered at Argonne. Reproduced with permission from Savina et.al.[190]

Writing on the sample case (Figure 5) indicated that the sample was first measured on March 3, 1948,

and again in 1976. Gamma and alpha spectroscopy, X-ray fluorescence, energy-dispersive X-ray spectroscopy, scanning electron microscopy, and resonant ion mass spectrometry were used to obtain information on elemental composition, plutonium isotopic analysis, and surface topography of the sample. Savina et.al discovered that the sample was nearly pure ^{239}Pu originating from 1948 with a surface morphology suggestive of electrodeposition. Given the time period and the purity, the sample was most likely derived from the X-10 reactor at Hanford [190]. This exercise showed that nuclear forensics is useful in simply identifying material with an unknown backstory.

c. Unknown Powder

In 2018, a yellow powder containing uranium (Figure 6), but with unknown origin and chemical composition, at Los Alamos National Laboratory (LANL) was analyzed by gamma spectrometry and micro-X-ray fluorescence to determine its chemical and isotopic composition [173]. Non-destructive gamma analysis determined that the sample was highly enriched in ^{235}U , approximately 96.5 wt%. However, micro-XRF indicated that the powder contained $\sim 90\%$ Nb and only 5% U. The yellow material was actually Nb_2O_5 , and small black particles interspersed within the material contained uranium. Though the origin is not known, a hypothesis has been put forth that the powder may have been ^{235}U -Nb-C_x, an early test reactor fuel.

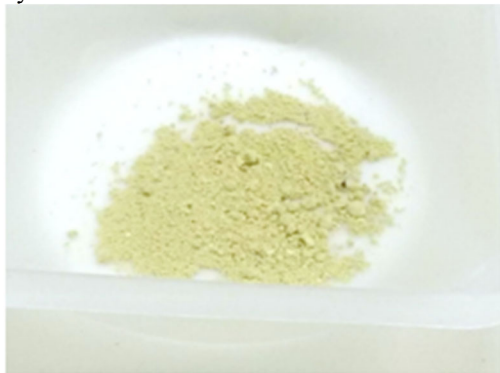


Figure 6. Sample of the unknown yellow powder. Reproduced with permission from Xu et.al.[173]

d. Summary

Although not as high-profile as international seizures, these case studies show that nuclear forensics is applicable to all levels of unknown radioactive material, from providing authentication of material to identifying an unknown material in storage.

Post-detonation Forensics

Nuclear forensics reviews generally focus on pre-detonation forensics, in which the radioactive material is found intact and analysis of the original material is possible via a wide variety of analytical techniques. Post-detonation forensics is the analysis of debris from an explosion, whether from a nuclear device or a radiological dispersal device (RDD). Analysis of debris focuses on fission products or isotopes from an RDD, but they often have to be separated from surrounding material (i.e. concrete, melt glass, urban materials) before they can be analyzed.

7. Radiological Dispersal Device

A radiological dispersal device is, at its simplest, a radiological source coupled with conventional explosives [191], the goal of which is to spread the source as far as possible with an explosion. Radioisotope sources include medical devices such as ^{60}Co and ^{137}Cs generators for radiotherapy, ^{90}Sr from radioisotope thermoelectric generators, and ^{192}Ir from industrial radiography equipment [191]. The use of an RDD would likely cause public panic, economic disruption, and loss of property use, rather than the physical devastation associated with a fission device. It is most likely, then, that an incident involving an RDD would occur in an urban environment, in order to spread the most panic. The majority of separation schemes for analysis of radiochemical isotopes have been developed for environmental samples based on nuclear testing, so the focus of current research on RDD isotopes (^{137}Cs , ^{90}Sr) is how to separate them from urban materials, such as concrete, steel, and other urban matrices [176,192-197].

a. Non-actinide Chronometers

Non-actinide radiochronometry has been a subject of interest for a number of years [55-58,60,198], and it is not generally mentioned in nuclear forensics reviews. Groups at Argonne National Laboratory (ANL) in the United States have focused on radiological dispersive devices (RDDs) in nuclear forensics, as they are more likely to occur than traditional nuclear weapon attacks [57]. Of particular concern are ^{90}Sr and ^{137}Cs which have been used in commercial applications such as radiotherapy and radioisotope thermoelectric generators (RTGs) in the United States and the former Soviet Union, and are more likely to be found out of regulatory control than traditional nuclear material. Age dating these materials can give scientists an idea of when the commercial isotopes may have been in use, and can help narrow down where it came from [57].

Strontium age-dating of radioactive material for forensics purposes is relatively new, with the first

procedure published in 2013 [59]. McLain et.al. were able to improve upon existing strontium-zirconium separation methods using Eichrom® DGA resin, achieving higher recoveries without the use of high concentrations of HF to keep Zr solubilized [57]. Labb et.al. discussed improving the separation of ^{137}Ba from Sr resin in order to age-date ^{137}Cs materials, which are commonly used in radiotherapy units and calibration sources [198]. A number of batch studies were done to show that Ba retention on Eichrom® Sr Resin decreases with increasing concentrations of tested chelating agents. Increasing the pH of the system also decreases the Ba retention. Barium recovery was improved by using chelating agents such as cyclohexyldiaminetetraacetic acid (CDTA) and nitrilotriacetic acid (NTA), which improves the likelihood of an accurate age determination of illicit radiological materials. Surrao et.al. published a similar study on ^{137}Ba and ^{90}Sr separation for ^{137}Cs and ^{90}Zr age determination [60].

b. Separations

An RDD is more likely to be detonated in an urban environment where it can do the most physical and psychological damage [197], rather than in a desert environment where nuclear tests have been conducted. The debris matrix is entirely different for an urban environment, and in the event of a detonation, the radiological isotopes would need to be separated from matrices such as steel, concrete, or cement [192,193,197,199]. Because ^{90}Sr and its daughter ^{90}Y are both pure beta emitters, quantification requires isolation from the debris matrix. McLain et.al have written a two-part series on the extraction of strontium from urban matrices such as steel, cement, and concrete [192,193]. Methods have been published on ^{90}Sr isolation using extraction chromatography resins, but only one applies to steel, and it requires significant effort to remove the remaining steel components prior to separation [197]. Additional difficulty stems from the fact that there are more than 3500 grades of steel which can contain differing amounts of metals such as aluminum, copper, chromium, manganese, and nickel [192]. The research by McLain shows that strontium can effectively be separated and analyzed using extraction chromatography, even in the presence of some common steel components, and that the extraction can be carried out without removing the steel constituents [192]. Other common matrices found in urban environments are concrete and cement [193]. The aggregate of materials used in creating concrete is extremely variable, and is often locally sourced. A representative aggregate material, dolomitic limestone, was chosen along with the two main types of cement. The primary constituents of

these materials are alkali and alkali-earth metals, which have a similar size and charge to strontium. The isolation of strontium in the presence of Ca, Cs, Na, Rb, Ba, and K, in addition to NH_4^+ , were investigated [193]. Significant differences in isolation of Sr were shown in experiments with high concentrations of Na, K, and Ca, which may necessitate a more complex separation scheme, such as those suggested by Maxwell, et.al [199].

8. Fission Device

After a fission induced nuclear event, such as those that were done during the nuclear testing era, the fission products can be used to determine the material in the original device, and therefore trace its origin. Both real debris, such as melt glass from nuclear tests [200-206], and surrogate debris made in the laboratory [123,207-216] are used in analysis. Various methods of dissolution and isolation of fission products from either melt glass matrices [217-221] or irradiated samples [212,222-225] have been recently studied to enrich the knowledge base of post-detonation forensics.

a. Source Preparations

Much like with pre-detonation forensics, reference materials, standards, and surrogate materials are useful for post-detonation analysis. Often, synthetic debris materials made with radioactive isotopes are synthesized and studied, due to the difficult nature of both obtaining and working with actual debris.

i. Real Debris

Trinitite is the most common debris material studied from an actual detonation [201,202,204,205,217]. Analyses ranging from high spatial resolution analysis [201] and ICP-MS [202,217] to isotope and element compositions [204,205] have been performed on Trinitite materials in recent years in order to determine the abundances of major and trace elements.

Donohue et.al. reported a study on comprehensive cross-sectional analysis on a variety of element abundances and $^{240}\text{Pu}/^{239}\text{Pu}$ ratios within Trinitite samples [201]. The data collected on the $^{240}\text{Pu}/^{239}\text{Pu}$ ratio indicated that residual fuel from the Trinity test was incorporated deeper into Trinitite than previously reported. This suggests a much greater disturbance of the local ground surface.

Bonamici et.al. used compositional analysis of post-detonation residues (i.e. Trinitite) to show how fallout composition is related to device composition [200]. Of particular interest were the major-element compositions of condensates, which form over a range

of temperature and pressure conditions. Major elements can be grouped based on their relative volatilities. However, more than 70 years after the Trinity explosion, radioactive isotopes are present only in trace quantities. Bonamici et.al were instead able to use stable elements to provide detailed information about fireball conditions.

ii. Synthetic Debris

It is difficult, if not impossible, to study short-lived fission and activation products using Trinitite due to its age. It can also be challenging to obtain real world debris samples, particularly in the post-nuclear testing era. Surrogate melt glass (the colloquial term for nuclear debris) is often made in the laboratory using isotopes (both radioactive and stable) added to a specific matrix, or by irradiation of a target material. Campbell et.al. synthesized and analyzed surrogate melt glass material using laser ablation (LA) ICP-MS [210]. Like McLain et.al., they were interested in creating a surrogate melt glass based on urban matrices such as steel, glass, cement, and its aggregates, and modeled after the World Trade Center debris. The composition of the melt glass was 50:50 soda lime (SiO_2 , CaCO_3 , and Na_2CO_3) and cement that contained silica sand, lime, limestone, iron, and clay. The final composition was doped with between 5 and 500 ppm of U_3O_8 with ^{235}U enrichment ranging from natural (0.72 %) to 80%. Scanning electron microscopy (SEM), X-ray photoelectron spectroscopy (XPS), and LA-ICP-MS were used to determine the morphology, bulk elements, and isotope ratios within the melt glass, respectively. The synthetic samples were similar to trinitite in morphology and chemical heterogeneity, and were used to assess the polyatomic interferences within the urban matrix that may interfere with trace constituent determination. The use of LA-ICP-MS, compared to solution phase ICP-MS, shows that the rapid LA technique has similar biases and statistical errors to the more complex solution phase analysis [210].

Many other teams, such as Molgaard et.al [207,213], Cook et.al [226], Liezers et.al [211,212], and Nizinski et.al [214] have published efforts to produce and characterize synthetic nuclear melt glass.

iii. Reference Materials

In another study, Biegalski et.al. activated post-detonation urban debris standards (NIST SRMs 4600 and 4601) using neutrons from a TRIGA reactor to identify nuclides of interest [224]. The NIST SRMs contain enriched uranium and a number of elements expected to be in post-detonation urban debris, including Al, As, Ca, Co, Cr, Cu, Fe, Mg, Mn, Mo, Ni,

Nb, P, K, Si, Sr, Sn, Ta, Th, Ti, W, V, Zn, and Zr. Analysis of the irradiated sample was done with gamma-ray spectroscopy. Over 140 peaks were identified in the spectrum, and were used to calculate the activity of each nuclide [224].

b. Separations

Similar to studies on RDD radionuclides, fission products from a nuclear event may need to be separated from the surrounding matrix to be effectively analyzed. Hubley et.al have studied ammonium bifluoride as a mechanism for the dissolution of trinitite and other post-detonation forensics samples [217-219]. Mason et.al, have used sonication assistance in conjunction with chemical dissolution [221]. Braysheer et.al used borate fusion to dissolve solid matrices, such as titanium dioxide, reference soils, concrete samples, and graphite [220]. These techniques need to be designed to dissolve a variety of matrices (i.e. steel, concrete, silica, and other refractory matrices), but also need to be rapid, and often field-deployable in order to quickly and accurately quantify nuclear debris [217].

Koeman et.al have been developing methodologies to effectively separate remnants of interest from post-detonation materials [227]. These remnants include metal components from the housing, electronics, or core material of the device, and are important for source attribution. Multiple matrices, including trinitite samples and synthetic melt glass, were characterized and treated with nitrogen trifluoride (NF_3). This method allows the separation of silica from minerals and glasses from non-volatile fluorinated species.

c. Summary

Though it is not as widely studied as pre-detonation forensics, mainly due to the fact that pre-detonation samples are historically more common, post-detonation nuclear forensics is an important topic for research. From identification and isolation of RDD isotopes from urban debris matrices, to the analysis of trinitite or surrogate melt glass samples for isotopic and elemental composition, post-detonation nuclear forensics is a complex field of study.

Conclusion

The field of nuclear forensics is inarguably imperative for the detection and attribution of both interdicted nuclear materials and the identification of nuclear samples of unknown provenance. The field has blossomed from the use of standard analytical procedures to recent developments in standards, morphology, radiochronometry, and international laboratory collaborations.

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